



Towards high performance broad spectral response fullerene based photosensitive organic field-effect transistors with tricomponent bulk heterojunctions



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ABSTRACT

Recently, broad spectral response phototransistors have drawn substantial attention due to their applications in the field of industry and science. However, it is difficult to synthesize appropriate photosensitive materials, which greatly limits their development. Integrating multicomponent bulk heterojunctions with high mobility materials to form hybrid planar-bulk heterojunction is a very effective approach to overcome these shortcomings. Herein, we reported broad spectral response photosensitive organic field-effect transistors with tricomponent bulk heterojunctions (Tri-BHJ) and high mobility channel transport layer, and they were sensitive over a bandwidth from ultraviolet–visible to near infrared. The Tri-BHJ composed of 3,4,9,10-perylenetetracarboxylic dianhydride (PTCDA), chloroaluminum phthalocyanine (AlClPc) and lead phthalocyanine (PbPc) was used as the photosensitive layer, fullerene (C₆₀) as the channel layer, and SiO₂ as the gate dielectric. By replacing SiO₂ with polyvinyl alcohol (PVA), the device performance was improved significantly, and the photosensitivity, photoresponsivity, external quantum efficiency and specific detectivity of the device built on PVA dielectric reached up to 10⁵, 108.44 A/W, 25325% and $\sim 2.7 \times 10^{12}$ Jones, respectively, which are comparable or even superior to those of commercially silicon and indium gallium arsenide photodetectors or other reported organic photodetectors. This work indicated new directions for the future development of high performance broad spectral response phototransistors.

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1. Introduction

Photodetectors capable of sensing light from the ultraviolet (UV)-visible (Vis) to the near infrared (NIR) region have received considerable attention in recent years due to their potential applications in industrial and scientific fields, such as image sensing, communications, military, medicine, environmental monitoring, remote control, security checks, day- and night-time surveillance,

chemical/biological sensing, spectroscopy, and industrial process control [1–8]. To date, commercial photodetectors have been mainly fabricated with inorganic semiconductors because of well-established infrastructures in manufacturing of inorganic devices [9]. However, inorganic photodetectors have some fundamental drawbacks, such as expensive cost and low temperature operation of some devices [10]. Moreover, much research on inorganic photodetector or materials are focused on different spectral region in the range from UV–Vis to NIR. For example, gallium nitride (GaN), zinc oxide (ZnO), silicon (Si) and indium-gallium-arsenide (InGaAs) are commonly used for sensing UV, Vis and NIR light, respectively [11–14]. Therefore, it is eagerly desirable to realize a facile photodetector with a broad response spectrum covered from UV–Vis to NIR. In this context, broad spectral response organic photodetectors have been introduced because of their advantages of low cost, lightweight, flexibility, large area, simple processing

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and room temperature operation [8,9,15,16]. In the past few years, broad spectral response organic photodetectors have aroused researchers' enormous interests. For instance, X. Gong et al. reported polymer photodetectors with spectral response from 300 nm to 1450 nm, showing a high detectivity greater than 10^{12} Jones [6]. H. Han et al. demonstrated broadband all-polymer phototransistors that achieved NIR sensing n-type and visible light sensing p-type transistor characteristics [9]. More recently, V. Q. Dang et al. have developed a flexible broad phototransistor based on methylammonium lead iodide perovskite-graphene hybrid channel [8]. Organic photodetectors can be classified into three types: organic photoconductor, organic photodiodes (OPDs) and organic phototransistors (OPTs) [17]. On account of the inherent advantages of high photosensitive and low noise, organic phototransistors are more favorable as broad spectral response organic photodetectors.

In general, broad spectral response photosensitive organic field-effect transistors (BSR-PhOFETs) can be realized by two strategies: (i) synthesizing new photosensitive materials with broad spectral absorption, such as perovskite [8,18]; (ii) mixed variety of photosensitive materials with different light absorption region, such methodologies including planar heterojunction [17], bulk heterojunction [9] and hybrid planar-bulk heterojunction [19]. For the first strategy, such materials need to have the capacity of molecular tailoring and the LUMO and HOMO energy levels tuning which will change the energy bandgap so that influences the absorption [20]. As a matter of fact, such materials are difficult to synthesize. On the contrary, the second strategy is simple and reliable to achieve broad spectrum detection. The integration of bulk heterojunction with broad spectral absorption and high mobility channel layer is expected to achieve high device performance. Regarding PhOFETs, up to now, most studies focus on the PhOFETs that are configured with a single layer or planar heterojunction [21–23]. Whereas, less attention has been paid to the PhOFETs with bulk heterojunction and hybrid planar-bulk heterojunction [9,24,25], of which benefits have been well demonstrated by our present papers which has been published [19]. So far, to the best of our knowledge, no study has been reported on the BSR-PhOFET with hybrid planar multi-component bulk heterojunction layers which can detect broad spectral lights from UV–Vis to NIR regions. In this work, we fabricated ultrahigh performance BSR-PhOFETs, in which tri-component bulk heterojunctions (Tri-BHJ) consisting of 3,4,9,10-perylenetetracarboxylic dianhydride (PTCDA), chloroaluminum phthalocyanine (AlClPc) and lead phthalocyanine (PbPc) was used as the photosensitive layer and fullerene (C_{60}) as the channel transport layer, and SiO_2 and polyvinyl alcohol (PVA) as the gate dielectric, respectively.

2. Experimental

2.1. Materials and device fabrication

All the organic materials were commercially obtained and used as received, except for AlClPc. PVA (99% purity, low molecular weight) was purchased from Alfa Aesars Ltd., C_{60} from Luminescence Technology Co. Ltd., PTCDA from J&K Chemical Ltd. and PbPc from Sigma-Aldrichs Ltd. AlClPc was synthesized and purified according to the procedure reported before [26,27]. The configuration of bottom-gate top-contact BSR-PhOFETs and the chemical structure of PTCDA, AlClPc, PbPc and C_{60} are shown in Fig. 1. Heavily doped n-type silicon (n^+ -Si) wafers with a 1100 nm thick thermally grown SiO_2 ($C_i = 3.14 \text{ nFcm}^{-2}$) were used as the gate and gate dielectric. For a comparison, indium-tin oxide (ITO) ($20 \Omega/\text{square}$) coated glass substrates were used as the gate and PVA as the gate dielectric. PTCDA:AlClPc:PbPc (weight ratio 1:1:1) bulk heterojunction (BHJ) was used as the photosensitive layer and C_{60} as the

channel layer. For the preparation of Si substrates, they were ultrasonically cleaned with piranha solution ($H_2SO_4:H_2O_2 = 7:3$) for 1 h, followed by acetone, ethanol, and deionized water for 10 min each and baked in a vacuum oven at 60°C for 20 min. For the preparation of ITO substrates, photoetching method was used to obtain a $5 \text{ mm} \times 20 \text{ mm}$ strip gate electrode. Subsequently, the substrates were ultrasonically cleaned with acetone, ethanol and deionized water for 10 min each and dried with blowing high-pure N_2 and baked in a vacuum oven at 60°C for 20 min. Then the PVA solution with a concentration of 20% in water was spin-coated at a speed of 3000 r/min onto ITO substrates. Whereafter, the PVA layer coated ITO substrates were annealed/cross-linked at 200°C for 2 h inside a vacuum oven to improve the PVA dielectric property, providing a uniform film with an approximate thickness of 1100 nm and a 4.24 nF/cm^2 capacitance. Next, 50 nm thick C_{60} films and 30 nm thick PTCDA:AlClPc:PbPc Tri-BHJ films were successively deposited onto the SiO_2 & PVA layers by vacuum thermal evaporation. After that, Au source-drain electrodes were thermally deposited on the top of the organic films via a shadow mask to define a $25 \mu\text{m}$ channel length and a 2 mm channel width. During all materials deposition, the chamber pressure was kept at a value of less than $8.0 \times 10^{-4} \text{ Pa}$ and the evaporation rate were maintained in the range of $0.10\text{--}0.2 \text{ \AA/s}$, monitored by a quartz crystal oscillator. Hereto, the SiO_2 -based and PVA-based BSR-PhOFETs were fabricated simultaneously, and all devices were deposited in the same batch.

2.2. Measurements and characterizations

All the measurements were carried out immediately after the devices were taken out of the thermal evaporation chamber, using an organic semiconductor characterization system in the dark and illumination conditions at room temperature. To measure the photosensitive effects, four commercially available UV–Vis–NIR lasers with a central wavelength of 405 nm (25 mW/cm^2), 532 nm (33 mW/cm^2), 655 nm (170 mW/cm^2), 850 nm (300 mW/cm^2) were used as the light source separately and different neutral density filters were exploited to vary the optical power density for illumination. For optical absorption measurements, the films of C_{60} (50 nm), C_{60} (50 nm)/PTCDA (30 nm), C_{60} (50 nm)/AlClPc (30 nm), C_{60} (50 nm)/PbPc (30 nm), C_{60} (50 nm)/PTCDA:AlClPc:PbPc (30 nm, weight ratio 1:1:1) were vacuum deposited on cleaned quartz substrates and measured by TU-1901 spectrometer, respectively. Crystal structure of C_{60} films were determined by X-ray diffraction (XRD) (Rigaku D/max-2400). Topography of organic channel layers were investigated by atomic force microscopy (AFM) (AFM, Agilent 5500) in the tapping mode.

3. Results and discussion

Fig. 2 shows the optical absorption spectra of C_{60} , C_{60} /PTCDA, C_{60} /AlClPc, C_{60} /PbPc and C_{60} /PTCDA:AlClPc:PbPc. The absorption of C_{60} /PTCDA, C_{60} /AlClPc and C_{60} /PbPc films are mainly in the range of 300–600 nm, 550–800 nm and 600–900 nm, respectively. Meanwhile, these active layers have different ratios of absorption to other range of spectrum. Moreover, C_{60} /AlClPc and C_{60} /PbPc films also have strong absorption in the range of 300–500 nm, which originates from not only C_{60} , but also AlClPc or PbPc film. It is observed that, the absorption of C_{60} /PTCDA:AlClPc:PbPc films covered a wide wavelength range of 300–900 nm (see green line in Fig. 2). Thereby, there exist mutual-complementary spectra for all the active materials in terms of hybrid planar tri-component bulk heterojunction, resulting in a complete coverage of UV–Vis–NIR photoabsorption. This affords the capability of broad spectral photodetection under the irradiation of UV–Vis–NIR light. It is

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